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Transport of sewage molecular markers through saturated soil column and effect of easily biodegradable primary substrate on their removal

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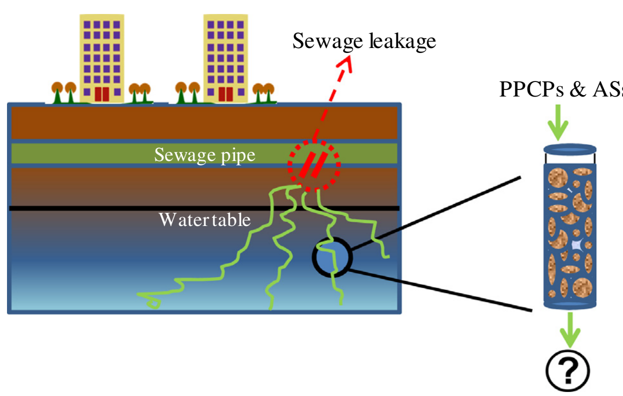
HIGHLIGHTS

- Transport of six PPCPs and three ASs through saturated soil column was determined.
- Transport of ACT, CF and DEET were limited by adsorption and biodegradation.
- ACF, CBZ, CTMT, CYC and SAC were recommended to be sewage molecular markers.
- Except CBZ and DEET, other compounds showed negative correlation by adding acetate.

GRAPHICAL ABSTRACT



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ABSTRACT

Pharmaceutical and personal care products (PPCPs) and artificial sweeteners (ASs) are emerging organic contaminants (EOCs) in the aquatic environment. The presence of PPCPs and ASs in water bodies has an ecologic potential risk and health concern. Therefore, it is needed to detect the pollution sources by understanding the transport behavior of sewage molecular markers in a subsurface area. The aim of this study was to evaluate transport of nine selected molecular markers through saturated soil column experiments. The selected sewage molecular markers in this study were six PPCPs including acetaminophen (ACT), carbamazepine (CBZ), caffeine (CF), crotamiton (CTMT), diethyltoluamide (DEET), salicylic acid (SA) and three ASs including acesulfame (ACF), cyclamate (CYC), and saccharine (SAC). Results confirmed that ACF, CBZ, CTMT, CYC and SAC were suitable to be used as sewage molecular markers since they were almost stable against sorption and biodegradation process during soil column experiments. In contrast, transport of ACT, CF and DEET were limited by both sorption and biodegradation processes and 100% removal efficiency was achieved in the biotic column.

Moreover, in this study the effect of different acetate concentration (0–100 mg/L) as an easily biodegradable primary substrate on a removal of PPCPs and ASs was also studied. Results showed a negative correlation ($r^2 > 0.75$) between the removal of some selected sewage chemical markers including ACF, CF, ACT, CYC, SAC and acetate concentration. CTMT also decreased with the addition of acetate, but increasing acetate concentration did not affect on its removal. CBZ and DEET removal were not dependent on the presence of acetate.

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1. Introduction

There is a concern about the effect of emerging organic compounds (EOCs) on groundwater and surface water quality and their adverse impacts on human health (Molander et al., 2009; Pomati et al., 2006; Sánchez-Avila et al., 2012). In the last few decades, fate and transport of pharmaceutical and personal care products (PPCPs) and artificial sweeteners (ASs) as two main groups of EOCs have been increasingly gaining attention (Chefetz et al., 2008; Li, 2014; Roy et al., 2014; Wolf et al., 2012). PPCPs are widely used in homes, hospitals and manufacturing which provide high opportunity to contaminate aquatic and land ecosystems via point sources such as treated wastewater effluent or from non-point sources such as illegal discharge, sewer overflow and sewer lines (Ellis, 2006; Liu and Wong, 2013). Beside PPCPs usage, ASs as a sugar substitutes are used in many products such as low calorie beverage (Tran et al., 2014; Van Stempvoort et al., 2011). The use of different kinds of medication and also sweeteners in all over the world plus rapid population growth and urbanization, cause the ubiquitous presence of PPCPs and ASs. The health effects of these chemicals are not clearly known, however, some of the chemicals and toxins may cause disruption in the human endocrine system (National Resources Defense Council and Environmental Integrity Project, 2004).

The fate and transport of EOCs in wastewater treatment plants (WWTPs) and soil aquifer treatment (SAT) have been investigated in previous studies (Arlos et al., 2014; Arye et al., 2011; Luo et al., 2014; Pal et al., 2010; Snip et al., 2014). Numerous studies reported on the presence of PPCPs and ASs in WWTP effluents (Kolpin et al., 2002; Lishman et al., 2006; Ternes, 1998; Yu et al., 2006). Therefore, WWTP effluents as a point source of PPCPs and ASs may affect aquatic ecosystems if it is released into the environment (Glassmeyer, 2005a,b; Maeng et al., 2011). Beside WWTP effluents, some non-point sources of EOCs such as leakage from sewer lines, illegal discharge, sewer overflows, wildlife and underestimated surface runoff may cause contamination of water bodies by PPCPs and ASs (Arslan-Alaton and Olmez-Hanci, 2013; Daughton, 2013; Díaz-Cruz and Barceló, 2008; Jurado et al., 2012; Kasprzyk-Hordern et al., 2008; Wolf et al., 2012). One of the important non-point sources of PPCPs and ASs into the environment is leaking sewage from domestic sewage pipelines either from cracks in pipes due to the pipe aging, corrosion and construction or from the inappropriate pipe junction (Tallon et al., 2005; Wolf et al., 2012). Leakage of untreated sewage to groundwater can be considered as a main cause of water quality degradation. Especially in tropical Asian regions, sewage pollution is a severe health risk to the people that live near rivers and waterways. Thus, there is a significance need to find suitable sewage molecular markers to detect the sewage leak and consequently eliminate groundwater and surface water contamination.

Previous studies suggested to detect sewer leakage by measuring the concentration of sewage molecular markers in nearby surface water and groundwater (Cronin et al., 2006; Kuroda et al., 2012; Tran et al., 2014). Some studies selected $PPCP_S$ and ASs as sewage chemical indicators (Aga et al., 2009; Andreozzi et al., 2003; Caliman and Gavrilescu, 2009; Henschel et al., 1997). Regarding to these studies, many of the broad lists have been developed for sewage markers which cause multiple potential markers, consuming a lot of time and also increasing high cost of analysis (Cronin et al., 2006; Eaton, 2012; Glassmeyer et al., 2005a,b). Thus, it remains very challenging to accurately estimate sewage leakage and to fully understand its migration in the subsurface area. To evaluate the sewage contamination of ground/surface water, monitoring the occurrence of these indicators in receiving water has been usually applied. However, it has been shown that chemical indicators can be lost by sorption to the soil and biodegradation by microbial activity. This means that the absence/presence of sewage indicators in receiving water bodies sometimes does not reflect the real impacts of sewage contamination on ground/surface water quality if other information on their transport is not provided. To eliminate these limitations, using soil column as a suitable tool to study transport behavior of sewage contaminants was recommended (Lewis and Sjöstrom, 2010; Magga et al., 2008). Soil column studies can provide some opportunities to determine transport mechanisms of sewage molecular markers through column length under different defined conditions. The most important transport mechanisms of sewage molecular markers in a subsurface area, are adsorption and biodegradation processes (Kagle et al., 2009; Mrozik and Stefańska, 2014).

Transport of sewage molecular marker may also be affected by various environmental factors which may increase or decrease their attenuation through a subsurface area. It is supposed that the presence of primary substrates at ppm level can provide enough carbon and energy sources for soil microbial community and consequently affect degradation of selected sewage chemical markers through the soil. The impact of primary substrates on removal of some EOCs through engineered aquifer recharge and filtration has been studied (Alidina et al., 2014; Onesios-Barry et al., 2014; Onesios and Bouwer, 2012), however there is no information on the role of biodegradable primary substrates on degradation of selected sewage chemical compounds through natural system.

Presumably tropical weather can have a special impact on the pollution transport through subsurface since most of the time the moisture content of the soil will be high and it will create the anoxic condition. Thus, the present study focused on saturated condition since in tropical regions, heavy rains elevate water level up and create saturation condition near sewage pipes. Furthermore, the risk and speed of spreading sewage contaminants into different directions is higher in a saturated area than unsaturated one.

This paper aimed to address the role of adsorption and biodegradation on a transport of selected sewage molecular markers in the subsurface by examining biotic and abiotic saturated soil columns. The selected sewage chemical markers included six selected PPCPs namely acetaminophen (ACT), carbamazepine (CBZ), caffeine (CF), crotamiton (CTMT), diethyltoluamide (DEET), salicylic acid (SA) and three ASs including acesulfame (ACF), cyclamate (CYC) and saccharine (SAC). The present study also evaluated the effect of different concentration of easily biodegradable primary substrate (supplied as CH_3COONa) on the removal of target chemical compounds. This study made a contribution to research on detecting sewage contamination in water bodies by demonstrating suitable sewage molecular markers based on the most important transport mechanisms such as adsorption and biodegradation data in a saturated soil column.

2. Materials and methods

2.1. Target molecular markers and tested soil

The selection of sewage molecular markers in this study was based on their physicochemical properties, high consumption and potential risk in aquatic ecosystems. One of the most important physicochemical properties of sewage molecular markers is hydrophilic characteristics (low log K_{ow}) in a subsurface area (Aga et al., 2009; Heberer, 2002). The physicochemical properties of the nine selected molecular markers are shown in Supplementary information A. Moreover, the selected sewage markers in present study are suggested by previous researchers